



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁵ : C08K 5/5399, 5/529, C08L 77/00 C08L 67/02, C08G 69/48, 63/91	A1	(11) International Publication Number: WO 93/24566 (43) International Publication Date: 9 December 1993 (09.12.93)
(21) International Application Number: PCT/US93/05281 (22) International Filing Date: 2 June 1993 (02.06.93) (30) Priority data: 892,844 3 June 1992 (03.06.92) US 893,184 3 June 1992 (03.06.92) US (71) Applicant: ALLIED-SIGNAL INC. [US/US]; 101 Columbia Road, P.O. Box 2245, Morristown, NJ 07962-2245 (US). (72) Inventors: VANBUSKIRK, Bruce ; 1 Fred Terrace, Dover, NJ 07801 (US). AKKAPEDDI, Murali, Krishna ; 7 Manor Drive, Morristown, NJ 07960 (US). GLANS, Jeffrey, Harper ; 20 Washington Avenue, Morristown, NJ 07960 (US).		(74) Agent: BLEEKER, Ronald, A.; Allied-Signal Inc., Law Dept. (C.A. McNally), 101 Columbia Road, P.O. Box 2245, Morristown, NJ 07962-2245 (US). (81) Designated States: AU, CA, JP, KR, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i>
(54) Title: PROCESS FOR PRODUCING POLYESTER OR POLYAMIDE COMPOSITIONS COMPRISING LACTAMYL PHOSPHITES AS CHAIN EXTENDING AGENTS (57) Abstract Polyester or polyamide compositions of intermediate and high molecular weights which are attained by reacting at least one lower molecular weight polyester or polyamide with an effective amount of a lactamyl phosphite. The reaction may be carried out by blending these constituents and any other optional constituents and is preferably formed by melt blending.		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	FR	France	MR	Mauritania
AU	Australia	GA	Gabon	MW	Malawi
BB	Barbados	GB	United Kingdom	NL	Netherlands
BE	Belgium	GN	Guinea	NO	Norway
BF	Burkina Faso	GR	Greece	NZ	New Zealand
BG	Bulgaria	HU	Hungary	PL	Poland
BJ	Benin	IE	Ireland	PT	Portugal
BR	Brazil	IT	Italy	RO	Romania
CA	Canada	JP	Japan	RU	Russian Federation
CF	Central African Republic	KP	Democratic People's Republic of Korea	SD	Sudan
CG	Congo	KR	Republic of Korea	SE	Sweden
CH	Switzerland	KZ	Kazakhstan	SK	Slovak Republic
CI	Côte d'Ivoire	LI	Liechtenstein	SN	Senegal
CM	Cameroon	LK	Sri Lanka	SU	Soviet Union
CS	Czechoslovakia	LU	Luxembourg	TD	Chad
CZ	Czech Republic	MC	Monaco	TG	Togo
DE	Germany	MG	Madagascar	UA	Ukraine
DK	Denmark	ML	Mali	US	United States of America
ES	Spain	MN	Mongolia	VN	Viet Nam
FI	Finland				

- 1 -

5

PROCESS FOR PRODUCING POLYESTER OR POLYAMIDE
COMPOSITIONS COMPRISING LACTAMYL PHOSPHITES
AS CHAIN EXTENDING AGENTS

10

BACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to polyester or polyamide compositions, more particularly to compositions and processes which are related to chain extended polyester or polyamide compositions.

2. Description of the Prior Art

Polyester and polyamide compositions are known as useful in a wide range of applications as engineering materials due to their favorable physical properties. See for example, US Patent 4,568,720; 5,037,897 and EP Published Application Nos. 0 232 878 and 0 232 879.

SUMMARY OF THE INVENTION

In one aspect of the invention, there are provided polyester compositions of intermediate and high molecular weights which are attained by reacting with at least one lower molecular weight polyester or polyamide with an effective amount of a lactamylphosphite as a constituent. The reaction may be carried out by blending these constituents and any other optional constituents in a melt. The lactamyl phosphate acts as chain extender to the polyesters and polyamides, and thereby providing materials which exhibit improved physical characteristics than those provided in the art.

- 2 -

Further aspects of the invention not particularly recited here will become apparent upon a reading of the accompanying specification of the preferred embodiments and the claims below.

5 **DESCRIPTION OF THE PREFERRED EMBODIMENTS**

Compositions of the present invention include the reaction product of one or more polyesters or one or more polyamides with one or more lactamyl phosphites.

10 Polyesters which may be employed in the process of this invention are linear or branched polyesters, which may be used singly or in mixture of two or more types of polyesters. The particular polyester or polyesters selected may be homopolyesters, copolyesters or mixtures thereof. Generally the polyesters used are the
15 condensation product of an organic dicarboxylic acid and an organic diol, which will be more particularly described below. The type of polyester is not critical and the particular polyester chosen for use in any particular situation will depend essentially on the ultimate physical properties of the final article of manufacture.

20 Suitable polyesters include those which are the condensation reaction products of aromatic, cycloaliphatic and aliphatic diols. Particularly suitable aromatic diols include those having from 6 to 12 carbon atoms, such as but not limited to:

 bis-(p-hydroxyphenyl)ether,
 bis-(p-hydroxyphenyl)thioether,
25 (bis-(p-hydroxyphenyl)-sulphone,
 (bis-(p-hydroxyphenyl)-methane),
 (bis-(p-hydroxyphenyl)-ethanes),
 1-phenol-(bis-(p-hydroxyphenyl)-methane),
 diphenyl-(bis-(p-hydroxyphenyl)-methane),
30 2,2-bis(4'-hydroxy'-dimethylephenyl)propane,

- 3 -

1,1- or 2,2-(bis(p-hydroxyphenyl)-butane),
1,1-dichloro-or 1,1,1-trichloro-
2,2-(bis-(p-hydroxyphenyl)-ethanes)
1,1 -(bis-(p-hydroxyphenyl)-cyclopentane)
5 2,2-(bis-(p-hydroxyphenyl)-propane (bisphenol A),
1,1-(bis-(p-hydroxyphenyl)-cyclohexane) (bisphenol C),p-
xyleneglycol,
2,5-dichloro-p-xylylene glycol,
p-xylene diol, and the like.

10 Useful cycloaliphatic diols include those from about 5 to about
8 carbon atoms, of which 1,4-dihydroxy cyclohexane, 1,4-dihydroxy
methylcyclohexane, 1,3-dihydroxy cyclopentane, 1,5-
dihydroxycycloheptane, 1,5-dihydroxycyclooctane, and 1,4-
cyclohexane dimethanol are suitable non-limiting examples.

15 Suitable aliphatic diols include but are not limited to diols of
from about 2 to about 12 carbon atoms, especially preferred being
those having from about 2 to about 6 carbon atoms. Examples of such
preferred diol precursors include 1,2-propylene glycol, 1,3-propylene
glycol, ethylene glycol, 1,8-octanediol, 1,4-nonanediol, neopentyl
20 glycol, pentyl glycol, 1,6-hexanediol, 1,4-butanediol, and the like.
Cycloaliphatic diols and aliphatic diols are preferred for use. Propylene
glycol, ethylene glycol, neopentyl diol, 1,4-dihydroxy
methylcyclohexane and 1,4-butanediol are particularly preferred.

25 Suitable dicarboxylic acids which find use as precursors in the
preparation of useful polyesters include linear and branched chain
saturated aliphatic dicarboxylic acids, aromatic dicarboxylic acids and
cycloaliphatic dicarboxylic acids. Polyesters containing up to about 80
% repeat units of monomer containing both hydroxyl and carboxylic
groups may also be used in the practice of this invention. Illustrative

- 4 -

aliphatic dicarboxylic acids which can be used in this invention are those having from about 2 to about 50 carbon atoms, such as oxalic acid, Malsonic acid, dimethylmalonic acid, succinic acid, octadecylsuccinic acid, pimelic acid, adipic acid, trimethyladipic acid, 5 sebacic acid, suberic acid, azelaic acid and dimeric acids (dimerization products of unsaturated aliphatic carboxylic acids such as oleic acid) and alkylated malsonic and succinic acids, such as octadecylsuccinic acid, and the like. Illustrative of suitable cycloaliphatic dicarboxylic acids are those having from about 6 to 10 about 15 carbon atoms, such as 1,3-cyclobutanedicarboxylic acid, 1,2-cyclopentanedicarboxylic acid, 1,3 and 1,4-dicarboxymethylcyclohexane and 4,4 -dicyclooxydicarboxylic acid, and the like.

Polyester compounds prepared from the condensation of a diol 15 and an aromatic dicarboxylic acid are preferred for use with this invention. Illustrative of such useful aromatic carboxylic acids are terephthalic acid, isophthalic acid, and o-phthalic acid, 1,3 -, 1,4-, 2,6 as well as 2,7-naphthalenedicarboxylic acid, 4,4-diphenyldicarboxylic acid, 4,4 -diphenylsulphone-dicarboxylic acid, 20 1,1,3-trimethyl-5-carboxy-3-(p-carboxyphenyl)-in Dane, diphenyl ether 4,4-dicarboxylic acid bis-p(carboxyphenyl)methane and the like. In the most preferred embodiments, poly(ethylene terephthalate) (herein sometimes referred to as "PET"), Poly(butylene terephthalate) (herein sometimes referred to as "PBT"), and Poly(1,4-cyclohexane dim 25 ethylene terephthalate) are the polyesters of choice, of which PET is the most preferred.

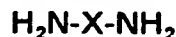
Polyamides which find use in accordance with the present invention include those which may be obtained by the polymerization of equimolar proportions of a diamine having two or more carbon 30 atoms between the amine terminal groups with a dicarboxylic acid, or

- 5 -

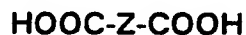
alternately that obtained by the polymerization of a monoamine carboxylic acid or an internal lactam thereof with an equimolar proportion of a diamine and a dicarboxylic acid.

Further, suitable polyamides may be derived by the
5 condensation of a monoaminocarboxylic acid or an internal lactam thereof having at least two carbon atoms between the amino and the carboxylic acid groups, as well as other means. General procedures useful for the preparation of polyamides are well known to the art, and the details of their formation are well described under the heading
10 ~ Polyamides ~ in the Encyclopedia of Polymer Science and Technology, published by John Wiley & Sons, Inc., Vol. 10, PPS. 487-491, (1969).

Suitable diamines include those having the formula



15 wherein n has an integer value of 1 - 16 and X is (CH₂)_n, or may further be such compounds as trimethylenediamine, tetramethylenediamine, pentamethylenediamine, hexamethylenediamine, octamethylenediamine, decamethylenediamine, dodecamethylenediamine,
20 hexadecamethylenediamine, aromatic diamines such as p-phenylenediamine, 4,4'-diaminodiphenyl ether, 4,4'-diaminodiphenyl sulphone, 4,4'-diaminodiphenylmethane, alkylated diamines such as 2,2- dimethylpentamethylenediamine, 2,2,4- trimethylhexamethylenediamine, and 2,4,4-
25 trimethylpentamethylenediamine, as well as cycloaliphatic diamines, such as diaminodicyclohexylmethane, and other compounds. The dicarboxylic acids useful in the formation of polyamides are preferably those which are represented by the general formula



30 wherein Z is representative of a divalent aliphatic radical containing at

- 6 -

least 2 carbon atoms, such as adipic acid, sebacic acid, octadecanedioic acid, pimelic acid, subeic acid, azelaic acid, undecanedioic acid, and glutaric acid. The dicarboxylic acids may be aliphatic acids, or aromatic acids, such as isophthalic acid and
 5 terephthalic acid.

By means of example, suitable polyamides include:

polypyrrolidone (nylon 4), polycaprolactam (nylon 6), polyheptolactam (nylon 7), polycaprylactam (nylon 8), polynonanolactam (nylon 9), polyundecaneolactam (nylon 11), polydodecanolactam (nylon 12),
 10 Polk(tetramethylenedioxamWe) (nylon 4,2), Polk(tetramethyleneadipamide) (nylon 4,6), Polk(tetramethylenediamine ~ o-isophthalic acid) (nylon 4,1), polyhexamethylene adipamide (nylon 6,6), polyhexamethylene azelaamide (nylon 6,9), polyhexamethylene sebacamide (nylon 6,10),
 15 polyhexamethylene isophthalamide (nylon 6,UP), polymetaxylylene adipamide (nylon MUD,6), the polyamide of n-dodecanedioic acid and hexamethylenediamine (nylon 6,12), the polyamide of dodecamethylenediamine and n-dodecanedioic acid (nylon 12,12), as well as copolymers thereof which include: hexamethylene adipamide-
 20 caprolactam (nylon 6,6/6), hexamethylene adipamide/hexamethylene-isophthalamide (nylon 6,6/61P), hexamethylene adipamide/hexamethylene-terephthalamide (nylon 6,6/IT), trim ethylene adipamide-hexamethylene-azelaamide (nylon trim ethyl,6/6), and hexamethylene adipamide-hexamethylene-
 25 azelaamide caprolactam (nylon 6,6/6,9/6) as well as others which are not particularly delineated here.

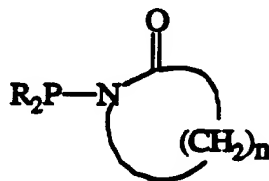
The polyamides may contain various terminal functionalities of which the most preferred are terminal functionalities which include both amino functional groups as one terminal functional group and a
 30 carboxyl group as another functional group; these may be present in

- 7 -

various relative proportions such as where the amount of carboxyl groups predominates over the amino functional groups. Preferred are those which are termed as "balanced", having an approximately equal proportion of amino functional groups to carboxyl functional groups.

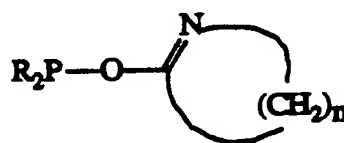
- 5 The polyamides may be of any desired molecular weight or molecular weight distribution but preferably having a number average molecular weight of from about 15,000 to about 40,000 as measured by end group titration. Preferred polyamides are nylon 66 and nylon 6.

The compositions according to the instant invention further include at
10 least one lactamyl phosphite which may be generally represented by the following formula[1]:



15

or alternately in accordance with formula [2]



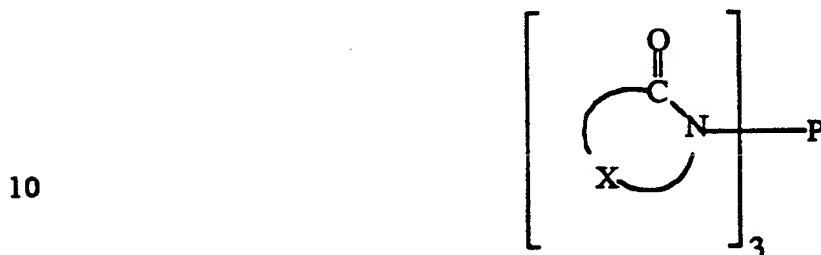
20

wherein the substituent group ERR may be any organic moiety such as an ethoxy, alkoxy, aryloxy, aryl, or alkyl group. Such lactamyl

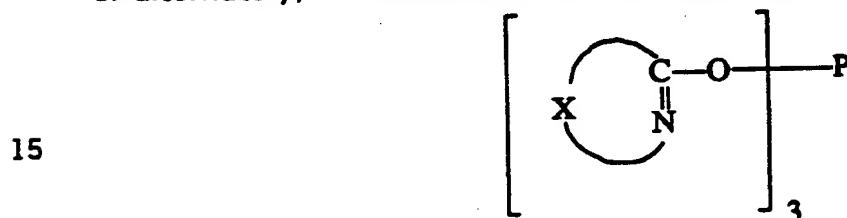
- 25 phosphites, especially wherein the substituent R is an ethoxy group is particularly described in an article by R. Mat Eva and N. Dencheva in the Journal of Polymer Science, Part A - Polymer Chemistry 30 1449 (1992). Further useful and preferred lactamyl phosphites are phosphoroustris lactams (sometimes hereinafter referred to as "RTCP") which find use in the present invention. These
- 30

- 8 -

phosphoroustrisactams are particularly described in US. Patent 5,118,805 for PHOSPHOROUSTRISACTAMS AND METHODS FOR THEIR PRODUCTIONS, and assigned to the same assignee as the instant application, the complete contents of which are herein incorporated by reference. The phosphoroustrisactams may be described as compounds in accordance with formula [3], below



or alternately, in accordance with formula [4],



where X represents a chain of CH₂ monomer repeat units of at least 1 and including up to 11 CH₂ monomer repeat units.

The phosphoroustrisactams may be produced for example by the reaction of a phosphoroustrihalide and a lactam, preferably caprolactam by the drop wise addition of the phosphoroustrihalide to the lactam in an aprotic organic solvent such as tetrahydrofuran, dioxane or glyme, and subsequently washed such as with water. The phosphoroustrisactams and their method of production is described more fully in commonly assigned US Patent 5,118,805 the contents of which are herein incorporated by reference.

The lactamyl phosphites, especially the phosphoroustrisactams utilized as a chain extending agent induce what is believed to be a coupling reaction between the hydroxyl end group of a polyester molecule or between the amino end group of a polyamide molecule

- 9 -

with the carboxyl end group of another polyester molecule with the carboxyl end group of another polyamide molecule, which results in a higher molecular weight polymer. Under ideal circumstances, equimolar amounts of hydroxyl or amino terminal groups and carboxyl terminal group are treated in the presence of a phosphoroustrislactam. The coupling reaction liberates as byproducts equimolar amounts of a lactam and phosphorous acid derivatives. Due to the relatively inert nature of these byproducts, the reacted polyester or polyamide, lactam and phosphorous acid derivatives may be removed by conventional methods. One beneficial feature of this reaction is that none of the byproducts of the coupling reaction are phenol comprising compounds, which are presently known to have toxic characteristics, which benefit is in contrast to the use of a tri(phenol phosphate) which forms phenol comprising compounds as a byproducts of its reaction.

What is to be understood by the term "effective amount" relative to the lactamyl phosphate is that an amount which is effective in providing sufficient coupling reactions between the terminal groups of the one or more polyesters and with the lactamyl phosphate which results in a chain extended polyester having a desired ultimate molecular weight. Such effective amounts may be readily determined experimentally and are to be understood to vary relative to the polyester or polyesters used and the ultimate molecular weight to be produced. Generally, the required amount of lactamyl phosphate is expected to be less than 10 parts by weight ("pbw") relative to 100 pbw of the polyester or polyesters used; preferably even less. Exemplary compositions are more particularly detailed in the Examples described below.

Other optional constituents which may be incorporated into the blends according to the instant invention include such materials as

- 10 -

fillers such as glass fibers or mineral fibers, impact modifiers, dyes, colorants, pigments, plasticizers, mold release agents, fire retardants, drip retardants, anti oxidants, UV stabilizing agents, mold release agents, colorants, anti static agents, nucleating agents, thermal
5 stabilizing agents, and the like. These optional constituents may be added to the mixture at any appropriate time during the production of the blend, and as they are well known to the art, are not here described with particularity. All of these optional constituents are commercially available.

10 The compositions according to the instant invention may be made by any technique or process, presently known or yet to be developed which will effect an intimate blending of the constituents of the compositions, particularly the polyester or polyamide, lactamyl phosphate and any additional constituents. By way of example, such
15 useful methods include formation of a solution in which the constituents are dissolved, suspended or dispersed in a suitable solvent, after which the solvent is removed from the resultant blend composition by conventional processes in order to form compositions in accordance with the teachings of the instant invention. An
20 alternative technique is by the dry blending the constituents in a dry particulate form, such as powders, pellets, flakes, pills or the like, and then heated to a temperature equal to or greater than the melting point of the highest melting constituent. A further variation on this technique which may be utilized where all of the desired constituents
25 are not available in powder form, is an additional process steps of mixing any liquid constituents or constituents in liquid form, subsequent to dry blending of the constituents, and thoroughly mixing the constituents, as well as removal of excess liquids during processing by well known techniques.

- 11 -

The polyester or polyamides may be reacted with the lactamyl phosphites in any manner which is effective for the chain coupling reaction to occur. Such methods require only that the lactamyl phosphites and the polyester or polyamide be well mixed and be at a sufficient temperature to allow for the coupling to occur. Exemplary techniques include batch wise fashions, or alternatively in continuous fashions. In the case of the former, a reaction vessel suitable to contain the constituents and to provide suitable reaction conditions, e.g. heat, temperature, adequate mixing of the constituents, atmosphere may be used, and such vessels include common laboratory glassware and flasks, Banbury mixers, and the like. In the case of the latter, an extruder of the single or multiple screw variety having at least one reaction zone may be utilized, as well as extruders having multiple zones, both in a series arrangement or in a parallel arrangement.

During production of compositions according to the instant invention, it is recognized that acceptable temperatures used in heating the constituents may vary over a wide range, and is dependent upon the constitution of any particular blend composition. Preferably, the temperature should be at least as high as the melting point of the polyester or polyamide but at the same time, should not be as high as the degradation temperatures of the polyester or polyamide. In particularly preferred embodiments, the temperature is such that the polyester will be retained in a molten state sufficiently long to allow for the lactamyl phosphite to react with polyester or polyamide and form a block or graft copolymer therewith.

The heating of the constituents may be carried out in any manner whereby the temperature constraints outlined above are achieved. In one contemplated method, the heating step is carried out at a temperature which is equal to or greater than the melting point of

- 12 -

the desired resultant composition. In an alternative method, the constituents are heated so that the temperature is increased as a function of time over the course of any heating process to cause the melting of constituents in the manner described in this specification, and to maintain this mixture in a molten state. Other methods not particularly described here, but which may be utilized in forming compositions according to the present invention are contemplated and considered within the scope of the invention.

Pressures are not contemplated to have any critical effect, and can be widely varied without adversely effecting the process of forming the inventive compositions. consequently, heating can be conducted at pressures below, at, or above atmospheric pressure. In preferred embodiments, at least a portion of the heating step is carried out at a reduced pressure so to allow the removal of any volatile constituents or by products.

The production of compositions may be conducted under normal atmospheric conditions, or in the absence of air. Alternatively, the production of compositions may be conducted in a controlled atmosphere, such as in the presence of an inert gas, such as argon, nitrogen, carbon dioxide, or other inert gas.

The time needed to react the constituents may vary over a wide range, and is recognized to be a factor of such effects as the polyester selected, additional constituents selected, the concentration of each of the constituents forming the composition, the temperatures to be used as well as the type of heating step used, as well as the type of reaction vessel and the manner of forming the composition. These are factors which are known in the art as effecting reaction times. In most instances, the reaction time will vary between from about 5 seconds up to about 25 hours, preferably, the reaction times vary between about 30 seconds to about 1 hour.

- 13 -

Preferably, the process of forming compositions in accordance with the instant invention includes a process step of removing any by-products of the reaction, as well as unreacted phosphate compounds. The methods used may be any conventional means which does not
5 adversely effect the composition formed. In preferred embodiments, all or part of the unreacted phosphate compounds are removed as it is believed that such removal enhances the effectiveness of the grafting process and improves the mechanical and other properties of the blend. Ideally, a composition where all of the unreacted phosphate
10 compounds are removed forms the most preferred embodiment of the invention, however it is concurrently recognized that complete removal is not always possible. Preferably, such techniques include formation of the blend under vacuum conditions or under reduced pressures, during any heating step.

15 The compositions of the instant invention are suitable for the formation of articles by subsequent molding or forming techniques, including but not limited to compression, injection, extrusion, as well as other techniques not particularly recited here, but which are nonetheless useful in forming formed articles therefrom. Particularly,
20 the controllable molecular weights of the chain extended polyester compositions taught herein which include blow molding operations, injection molding techniques and in extrusion production techniques.

While not wishing to be bound by any theory, it is hypothesized by the inventors that the present invention provides a method of
25 effecting chain extension of one or more polyesters or polyamides which is attained by reacting one or more polyesters with an effective amount of a lactamyl phosphite by control of the process conditions of the reaction, a wide range of molecular weights may be attained. Further, when two or more dissimilar polyesters are reacted with the
30 lactamyl phosphite, especially phosphoroustris lactam, (hereinafter

- 14 -

sometimes referred to as ("RTCP"), a compatible polyester blend is attained.

The foregoing invention will be more apparent by reference to specific embodiments which are representative of the invention. It is
5 nonetheless to be understood that the particular embodiments described herein are provided for the purpose of illustration, and not be means of limitation, and that it is to be further understood that the present invention may be practiced in a manner which is not exemplified herein without departing from its scope.

10

EXAMPLES

In the following embodiments of the invention, it is to be understood that in the description of any composition, all percentages associated with a constituent used to form a composition are to be
15 understood as to be "percentage by weight" of the particular constituent relative to the composition of which it forms a part; exceptions to this convention will be particularly noted.

COMPARATIVE EXAMPLE 1

20 A poly(ethylene terephthalate) homopolymer which may be characterized as having an intrinsic viscosity of 0.7 as measured in phenol/TCE at 25 Del.C. The PET homopolymer was determined to exhibit a reduced viscosity of 0.66 as measured in phenol/TCE at 25 Del.C. The PET homopolymer was also determined to have 0% in
25 solubles.

EXAMPLE 1

A poly(ethylene terephthalate) homopolymer characterized as having an intrinsic viscosity of 0.7 as measured in phenol/TCE at 25
30 Del.C was reacted with 1 % of phosphoroustris lactam as prepared

- 15 -

according to US Patent 5,118,805 and generally described above. The PET and the TCP were reacted by melt blending the same in the TW-100 extruder as described above.

The reacted PET was determined to exhibit a reduced viscosity of 0.88 as measured by indicating that an increase in the molecular weight had been achieved, and further was found to have 0% in solubles which indicated that no appreciable amount of cross linked products were formed but rather, that linear chain extension was accomplished.

10

EXAMPLE 2

For each of the following examples the following materials were utilized. "Nylon 1" was a low to intermediate molecular weight Nylon 6 homopolymer resin which may be characterized as having a formic acid viscosity ("FAV") of 56 at 25 Del.C and about 9.5% extractable. "Nylon 2" was a commercially available grade of nylon 6 molding resin marketed by Allied-Signal Inc. as Cap Ron (R) 8209F which may be characterized as a high viscosity Nylon 6 film grade resin having a specific gravity of 1.13 according to ASTM D -792, a melting point of 420 Del.F. "Nylon 3" was a commercially available grade of Nylon 6 homopolymer molding resin marketed by Allied-Signal Inc. as Cap Ron (R) 8207F and sold as a film grade resin which may be characterized as a medium viscosity homopolymer Nylon 6 having a specific gravity of 1.13 and a melting point of 420 Del.F according to ASTM D-792. "TCP" was phosphoroustris lactam produced in accordance with US Patent 5,118,805. Each of the compositions were produced in accordance with the following general technique. Weighted amounts of the constituents were tumble blended together in a sealed container

- 16 -

to assure a homogeneous blend, after which each of the mixtures were supplied to the feed hopper placed at the throat of a Hake-Buechler Instrument Co. model TWO-100 twin screw vented extruder having two counter-rotating conical inter meshing mixing screws. The
5 extruder barrel had the temperature profiles and operating conditions noted on Table 1 below.

In the Table 1, the compositions of this invention are compositions 1, 2 and 3 which include the lactanyl phosphite, phosphorous-trislactam and for comparative compositions are
10 compositions A, B, C and D which do not include any lactanyl phosphite.

TABLE 1

<u>Composition</u>	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>	<u>1</u>	<u>2</u>	<u>3</u>
Nylon 1	100			100			
Nylon 2		100					
Nylon 3			100		99	98	97
phosphorous-trislactam	-	-	-	-	1	2	3
Extruder Temp. in deg. C							
zone 1	240	244	239	200-211	242	239	241
zone 2	258	259	258	230-233	259	262	262
zone 3	275	272	269	250-266	268	267	271
zone 4	280	280	280	250	280	280	280
die	275	270	269	250-272	270	273	275
speed, rpm	8787	87	87	50	87	87	87
pressure, psi	0-200	75-130	40-170	60	500-900	1200-2100	2200-2500
torque, m-gm	100-1500	2700-3500	220-2800	600	2800-4100	3800-4850	4700-5700

TABLE 1 - CONTINUATION

Composition	4	5	6	7	8	9	10	11
Nylon 1				99	98	97	99	99
Nylon 2	99	98	97					
Nylon 3								
phosphorous-trisilactam	1	2	3	1	2	3	1	1
Extruder Temp. in deg.C								
zone 1	247	247	245	241	243	243	200-210	200-211
zone 2	264	262	259	262	264	264	230-233	230-233
zone 3	268	271	264	272	272	272	250-266	250-266
zone 4	280	280	280	280	280	280	250	250
die	271	276	272	267	258	273	250-272	250-272
speed, rpm	87	87	87	88	88	88	50	50
pressur , psi	950-1100	1700-2300	700-2200		450-670	950-1130	280	220
torque, m-gm	5300-6200	6200-7400	5300-7300	2300-3500	3850-4300	3500-3800	1500	2500

- 19 -

The extrudate exiting the die was in the form of strands having a diameter of 1/8 inch, and were quickly passed into a water bath to quench and cool the strands. The strands were subsequently pelleted to form a feed stock useful for injection molding. A portion of the pellets of selected compositions were used to determine the physical properties thereof. The reduced viscosity was determined in m-cresol according to conventional techniques at 25 Del.C, the melt index was determined using the protocol of ASTM D-1238, the percent in solubles was determined by filtering the dissolved polymer through a screen and then drying and weighing the residue, and the formic acid viscosity ("FAV") was estimated using empirical techniques. The results are denoted on Table 2. Comparison of the results obtained from the compositions of this invention 1, 2 and 3 as compared with comparative compositions A, B, C and D indicates that appreciable increases in the viscosity of the compositions of this invention without an concurrent increase in the percent in solubles suggests that the resultant experimental compositions of this invention had been linearly chain extended as opposed to grafted or cross linked which would have manifested itself in a more dramatic increase in the percent insolubles.

TABLE 2

Compositi n	A	B	C	D	1	2	3
Reduced Viscosity in m-cresol	1.66	2.3	1.86	1.51	3.63	4.16	4.14
Melt Index*	8.122	1.42	4.415		no flow		
% Insolubles	0	<1	0	0	<1	<1	<1
FAV**	60	75	140	--	265	350	345
* Melt Index of 1000g sample at 235 deg. C							
** FAV is formic acid viscosity							
C mpositi n	4	5	6	7	8	9	10
Reduced Viscosity in m-cresol	3.71	4.53	3.82	2.7	3.09	3.28	2.55
Melt Index*		no flow			0.403		
% Insolubles	<1	<1	<1	<1	<1	<1	<1
FAV**	290	390	300	170	215	245	
							11
							2.31

- 21 -

WHAT WE CLAIM IS:

1. A composition comprising:
a polymer selected from the group consisting of polyamide and a polyester, and a lactamyl phosphite.
2. A composition according to claim 1 wherein said polymer is a polyester.
3. A composition according to claim 2 wherein the polyester is selected from the group consisting of poly(ethylene terephthalate) and poly(butylene terephthalate).
4. A composition according to claim 1 wherein said polymer is a polyamide.
5. A composition according to claim 4 wherein said polyamide is selected from the group consisting of nylon 6 and nylon 6,6.
6. A composition according to claim 5 wherein said polyamide is nylon 6.
7. A composition according to claim 1 wherein the lactamyl phosphite is a phosphoroustrislactam.
8. A composition according to claim 1 wherein the lactamyl phosphite comprises up to 10 parts by weight relative to 100 parts by weight of the polymer.
9. A process for producing chain extended polyester or polyamide compositions comprising the process step of:
reacting one or more lactamyl phosphites with one or more polyesters or polyamides.
10. The process according to claim 9 wherein the one or more lactamyl phosphites are one or more phosphoroustrislactams.

INTERNATIONAL SEARCH REPORT

International Application No.

PCT/US 93/05281

I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) ⁶		
According to International Patent Classification (IPC) or to both National Classification and IPC		
Int.Cl. 5 C08K5/5399; C08G69/48;	C08K5/529; C08G63/91	C08L77/00; C08L67/02
II. FIELDS SEARCHED		
Minimum Documentation Searched ⁷		
Classification System	Classification Symbols	
Int.Cl. 5	C08G ; C08K ; C08L	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁸		
III. DOCUMENTS CONSIDERED TO BE RELEVANT⁹		
Category ¹⁰	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³
A	WO,A,9 202 579 (ALLIED -SIGNAL INC) 20 February 1992 & US,A,5 037 897 cited in the application ---	1-3,7-8
A	SOVIET INVENTIONS ILLUSTRATED Section Ch: Chemical, week 8423, 18 July 1984, Derwent Publications Ltd. (London, GB) & SU,A,1 041 546 (PHYTOPATHOLOGY RES INST) 15 September 1983 see abstract --- -/--	1
<p>¹⁰ Special categories of cited documents : ¹⁰</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"A" document member of the same patent family</p>		
IV. CERTIFICATION		
Date of the Actual Completion of the International Search 12 AUGUST 1993		Date of Mailing of this International Search Report 25.08.93
International Searching Authority EUROPEAN PATENT OFFICE		Signature of Authorized Officer Kathryn Y.-P. Miao

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)

Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No.
A	JOURNAL OF APPLIED POLYMER SCIENCE vol. 34, no. 8, December 1987, NEW YORK, NY, USA pages 2769 - 2776 H. INATA, ET AL 'Chain Extenders for Polyester. VI. Properties of the Polyesters Chain-Extended by 2,2'-Bis(4H-3,1-Benzoxazin-4-one)' ---	
A	JOURNAL OF APPLIED POLYMER SCIENCE vol. 30, no. 8, August 1985, NEW YORK, USA pages 3325 - 3337 H. INATA, ET AL 'Chain Extenders for Polyesters. I. Addition-Type Chain Extenders Reactive with Carboxyl End Groups of Polyesters' ---	
A	CHEMICAL ABSTRACTS, vol. 76, no. 6, 7 February 1972, Columbus, Ohio, US; abstract no. 25774n, page 16 ;column 1 ; & JP,B,71 031 871 (TOYO SPINNING CO., LTD.) 17 September 1971 ---	
A	CHEMICAL ABSTRACTS, vol. 75, no. 2, 12 July 1971, Columbus, Ohio, US; abstract no. 7271w, page 64 ;column 1 ; & JP,B,70 030 950 (YUNICHIKA CO., LTD.) 7 October 1970 -----	

US 9305281
SA 75283

12/08/93

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO-A-9202579	20-02-92	US-A- 5037897 US-A- 5070152	06-08-91 03-12-91
